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## EROSION RESISTANCE EVALUATION OF POTENTIAL GUN BARREL COATINGS

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<p>The erosion resistance of tungsten/carbon and tantalum/tungsten deposited on AISI 4340 steel and molybdenum substrate were investigated. Erosion data ac- quired with the ARRADCOM test device indicated poor adhesion between each re- fractory metal coating and 4340 steel. Mechanical failure of the molybdenum substrate complicated the erosion data, but it was concluded that the tungsten/ carbon coating performed better than steel at higher pressures. No apparent ad- vantage was observed with the tantalum/tungsten coating at any pressure.</p>		

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20. ABSTRACT (cont)

Microscopic examination of the coatings showed extensive gas erosion of the surface of the tantalum/tungsten coating, but very little erosion in the tungsten/carbon surface.

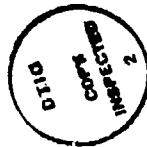
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## INTRODUCTION

This report documents an attempt to minimize or reduce gun barrel erosion in large caliber guns. The correction of the gun barrel erosion problem can be addressed from several directions. One approach is to formulate new propellants or modify current propellants to produce less erosive combustion products without significantly sacrificing propellant energetics; another is to modify gun barrels in some way to better resist the erosive combustion environments produced by current propellants. Both approaches have been used with varying degrees of success, including new low flame-temperature propellants and traditional plating of gun barrels with chrome.

Although refractory metals that are potentially superior to chromium have been available, and more are being discovered every day, limited use has been made of these materials because of the difficulties encountered in coating gun barrels. Recently, however, coating deposition technology has made such strides that use of these exotic refractory metals in gun barrels is now feasible. Specifically, Cullinan and D'Andrea<sup>1</sup> reported encouraging results in using tantalum coated 20-mm steel liners.

The main thrust of this program was to develop refractory coatings suitable for use in large caliber guns. Three candidate refractory metal alloys, tungsten/carbon alloy; (CM500) low-temperature deposited tungsten/carbon alloy; (CM500L), and 98% Ta/2% W alloy were applied to AISI 4340 steel and molybdenum substrates to test each alloy's erosion behavior under controlled, simulated gun firing conditions.

## PROCEDURE

### Tungsten/Carbon Coatings

Both AISI 4340 steel and molybdenum inserts were coated with tungsten/carbon alloy by means of the chemical vapor deposition (CVD) process. The steel samples were coated at low temperature (725 K) with tungsten carbon alloy (W/C), identified by the contractor as CM500L, while the molybdenum inserts were coated at higher temperatures ranging from 1165 to 1245 K with the W/C alloy identified by the contractor as CM500.

To protect the steel substrate from the corrosivity of the reactive gases at the deposition temperature when  $WF_6$  is used as the tungsten precursor, an initial

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<sup>1</sup> R. Cullinan and G. D'Andrea, "Erosion Studies of Tantalum Gun Barrels," 1980 JANNAF Propulsion Meeting, vol I, p 201, 1980.



thin layer of relatively inert metal, in this case electroless nickel, was deposited onto the steel substrate. In addition to being inert, nickel is also compatible with both steel and tungsten. The deposition parameters, substrate temperature, chamber pressure, gas flow rate, and gas composition are presented in table 1.

#### Tantalum/Tungsten Coatings

Test inserts of AISI 4340 steel and molybdenum were coated with Ta/W alloy at deposition temperatures ranging from 1300-1450 K. An initial layer of titanium was deposited from the vapor phase onto the steel substrate to form an inter-layer which is compatible with both iron (steel) and tantalum. Titanium was selected rather than nickel because, in addition to the higher melting point, titanium also reacts with the carbon and the steel substrate to form a thin surface layer of titanium carbide (TiC). This stable TiC layer is an effective carbon diffusion barrier which prevents any further interaction of carbon in the steel with the tantalum coating. This eliminates the possibility of the formation of the undesired brittle TaC. After this titanization process, Ta/W alloy was coated onto the Ti-TiC layer. This was accomplished by passing chlorine over 90 Ta/10W alloy chips from a mixture of metal chloride gases,  $\text{TaCl}_5$  (g),  $\text{WCl}_6$  (g),  $\text{WCl}_5$  (g) which was reduced onto the substrate surface by hydrogen as Ta/W alloy. Although the target composition was 90% tantalum and 10% tungsten, the achieved composition was 98% tantalum and 2% tungsten. The working parameters for this process are presented in table 1.

#### Erosion Testing

The erosion data were obtained with a closed bomb modified to accept a gun barrel and a cylindrical metal erosion sleeve. This variation of the closed bomb is usually referred to as the vented erosion tester and is shown in figure 1. In this investigation, the erosion sleeves consisted of AISI 4340 steel, refractory metal coated AISI 4340 steel, and refractory metal coated molybdenum. Each sleeve was fabricated to have an outer diameter of 2.70 cm, an inner diameter of 0.95 cm, and a length of 2.06 cm. The average masses of the steel and molybdenum erosion sleeves were 80 g and 103 g, respectively. A pressure transducer positioned inside the chamber (closed bomb) was connected to a Nicolet digital oscilloscope which was scaled to display pressure versus time. To control pressure, a stainless steel rupture disc was inserted between the barrel and the erosion sleeve. In addition, the barrel was filled with water and a rubber stopper inserted into the muzzle to insure proper pressure buildup.

The Bach CM500L coated steel sleeve and the CM500 coated molybdenum sleeve were cleaned, weighed, fired three times, recleaned, and reweighed. After three shots, the average weight loss was used as a measure of erosivity. For the Ta/W-coated steel sleeves, an initial single-shot mass loss was measured for each sample. All subsequent measurements made on these samples were based on average

weight losses for three successive shots. All erosion measurements made on Ta/W-coated molybdenum sleeves were based on single shot weight-loss measurements.

The internal ballistics were controlled by arbitrarily adjusting the propelling charge weights to 30, 40 and 50 g which yielded peak pressure in the ranges of 150, 180 and 260 MPa, respectively. Since the same propellant (M30) was used throughout this study, flame temperature was a constant while burn times and peak pressures were measured charge-mass dependent parameters. The composition and physico-chemical properties of M30 propellant are listed in table 2.

## RESULTS AND COMPARISONS

### Steel Sleeves (AISI 4340)

#### CM500L Coatings

Two steel sleeves identified as nos 398 and 400 were received from the contractor with CM500L coatings of 0.010 cm and 0.015 cm thicknesses, respectively. cursory visual examination by the Large Caliber Weapon Systems Laboratory indicated that each coating was deposited uniformly along the bore surface with no apparent local areas of defects. However, the contractor reported<sup>2</sup> that the coating on sleeve no. 400 did not appear to be as well bonded to the bore surface as did the other coatings. The hardness of the coating on sleeve no. 398 varied from 700 kg/mm<sup>2</sup> HV<sub>500</sub> near the steel substrate to 2400 kg/mm<sup>2</sup> HV<sub>500</sub> at the i.d. free surface. This hardness gradient was caused by the incorporation of carbon into the tungsten coating during the process. This results in a carbon rich tungsten carbide and tungsten layer at the i.d. free surface which displays greater hardness.

The erosion test data for the CM500L-coated sleeves are presented in table 3. In this work, all bore surface losses are reported as volume losses rather than mass losses. This departure from the usual convention of reporting mass losses was necessary to normalize all erosion data to a common scale to make erosion comparisons among the different coatings and steel meaningful.

Erosion data, obtained in the 150 MPa range indicated that sleeve no. 398 with the 0.010 cm CM500L coating retained 89.0% of the coating after three shots and 73.7% after the ninth shot, while the sleeve no. 400 with the 0.015 cm coating retained only 0.78% of the coating after three shots and 0.00% after the six shots. On an average per shot basis, the sleeve with the 0.010 cm coating lost  $0.0017 \pm .00041 \text{ cm}^3$  and the other sleeve lost  $0.01045 \pm .01696 \text{ cm}^3$ . Steel sleeves on the other hand, displayed an average per shot erosion value of  $0.00067 \pm .00014$  in the 150 MPa pressure range (table 4). A comparison of these data suggested that 4340 steel apparently erodes less per shot than does CM500L alloy. The larger standard deviations observed in the erosion data for CM500L

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<sup>2</sup> Chemetal letter from Robert Holzl dated October 8, 1979.

coatings suggested that different phenomena may be involved in each erosion event. In contrast, the small standard deviations observed for the steel sleeves at all pressures indicated that the same phenomena (or phenomenon) occur in each erosion event.

Visual examination of each sleeve after erosion testing revealed that little coating was left on sleeve no. 400 while a substantial amount of coating remained on sleeve no. 398. As a result of poor bonding or adhesion between the CM500L alloy and the steel substrate, most of the coating was blown off sleeve no. 400 within the first three shots. Also, the erosion data acquired with the remaining shots reflected the combined erosion effects of the exposed steel substrate and the remnants of CM500L coatings.

Scanning electron microscope (SEM) photographs of the etched cross section of a CM500L coated sleeve, not previously erosion tested, are presented in figure 2. These photographs clearly illustrate the concentric coherent layers of steel, nickel, columnar tungsten, and the outer layer of tungsten/tungsten carbides. The layer containing the tungsten carbides also exhibits a myriad of worm holes which probably result from the polishing operation used in sample preparation. During polishing, particles of  $W_2C$  are pulled out of the matrix which scratch or gouge the surface of the matrix.<sup>2</sup> Examination of these worm holes indicated high carbon concentrations in the worm hole edges. A more highly magnified rendition of the coating shows the fine microstructure of the tungsten carbide layer. The SEM photographs of the CM500L coating remnants in figure 3 reveal the presence of cracks and wear tracks in coating after erosion testing. The wear tracks result from the erosive action of flowing hot gases, while the cracks may be a consequence of a structure-stress relationship. The growth morphology of the stressed tungsten layer is columnar which produces a microcrack network along grain boundaries that extends from the tungsten carbide surface to the steel substrate surface. Thermal-mechanical stresses produced during erosion testing adversely affected this microcrack network resulting in portions of this tungsten undercoating along with its tungsten carbide overlayer to break away from the steel substrate. This phenomenon was observed by Gibson<sup>3</sup> for chromium coatings exhibiting columnar morphology.

#### Tantalum/Tungsten Coatings (Steel Sleeves)

Steel sleeves coated with 0.10 cm and 0.019 cm of Ta/W alloy were received from the contractor. The hardness of these coatings ranged from 350 to 400 kg/mm<sup>2</sup> HV<sub>500</sub>. Energy dispersive x-ray analysis of the coatings revealed that the alloy composition was 98% Ta and 2% W, rather than the target composition of 90% Ta and 10% W. Hence, the hardness of the coatings was lower than expected. No titanium could be detected at the interface between the steel and the Ta/W alloy which indicated: (1) The steel substrate was not titanized as desired, and (2)

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<sup>3</sup> I. Gibson, "Development of A Vented Chamber Erosion System for Gun Barrel Coating Assessment Studies," (in press).

the Ta/W alloy was bonded directly to the steel substrate. Visual examination of the coatings, however, indicated apparent good interlayer bonding.

The Ta/W coated steel erosion data in table 5 were obtained in the 180 MPa pressure range. Sleeve no. 107 lost less than 1% during the first shot but lost the remainder of the coating during the next three shots. The other sleeve lost about 17% of the coating during the first shot and the remainder during the next three shots. Visual examination of each sleeve revealed massive spalling occurred during testing. In both cases, the erosion per shot level apparently exceeded that of 4340 steel. An SEM photograph presented in figure 4 clearly shows: (1) The erosion effects on the surfaces of the steel and the Ta/W deposit, (2) the columnar morphology, and (3) the attendant microcrack network. A more highly magnified view of the severely eroded steel substrate immediately adjacent to the same remnant Ta/W coating (fig. 5) illustrates the drastic erosion caused by gas flow eddy currents.

## Molybdenum Sleeves

### CM500 Coatings

The hardness of the CM500 coated samples ranged from 2100 to 2400 HV<sub>500</sub>. In contrast to the CM500L coatings; however, no hardness gradient was observed in the CM500 coating. This was considered indicative of a homogeneous coating in which all the alloy components were uniformly mixed in a single layer. The homogeneity of the fine microstructure of the CM500 coating is verified in the SEM photographs presented in figure 6.

The CM500 erosion test results, presented in table 5, were complicated by the fact that the molybdenum sleeves cracked longitudinally in eight out of ten cases. In these cases, the reported erosion values may be artificially inflated by the material lost during crack formation. In one case, sleeve no. 3, the cracking was so severe that it split in half. Examination of this sample revealed that, in addition to gross longitudinal cracks, small local cracks were evident in the coating. SEM photographs of a typical local crack, presented in figure 6, show that the crack in the coating terminates at the substrate interface, and that considerable gas erosion occurs at the substrate interface. Despite this catastrophic fracture of the coating and the cracking of the sleeve itself, CM500 displayed both excellent adhesion to the molybdenum substrate and good erosion resistance (fig. 7). It should also be noted that the leading edge of the bore surface of each sleeve exhibited extensive chipping. SEM photographs of a typical chipped leading edge presented in figure 8 clearly show a fractured coating with angular protuberances.

The erosion results presented in table 6 indicate that in the 150-MPa range, CM500 erosion performance is marginally worse than 4340 steel itself, but this may be an artifact caused by sleeve cracking and chipping. In the higher pressure regime, CM500 apparently performs better than steel. Also, the level of CM500 erosion does not increase significantly with erosion.

### Tantalum/Tungsten Coatings (Molybdenum Sleeves)

Single shot erosion data for Ta/W-coated molybdenum sleeves are listed in table 7. Again, these results have been negatively affected by longitudinal cracking of the molybdenum sleeves and by chipping of the coating on the leading edge of the bore surface. Only two out of six samples exceed or replicate the average per-shot erosion performance of steel at 150 MPa, but this may not reflect the true erosion characteristics of this coating. At higher pressures, only one sample matches the average per shot erosion performance of steel. Examination of the standard deviations and each single shot erosion datum suggest multi-phenomena effects may randomly occur. For those shots where the coating matches or outperforms steel, erosion is the dominant or only effect; all other shots probably include losses which result because of crack formation and edge chipping.

### CONCLUSIONS

From the data obtained, the following was concluded:

1. Tantalum/tungsten coatings offer little apparent advantage over steel in the ARRADCOM test device.

2. The tungsten/carbon alloy, CM500, is a candidate coating to reduce wear in gun tubes.

Adhesion problems with steel, however, have to be solved before this coating can be tested in a gun.

Table 1. Typical conditions for depositing CM500L and 90Ta-10W alloy

Deposit	Substrate	Gas flow rates (ml/min)							Chamber pressure (MPa)	Substrate temp (K)	Run time (min)
		H <sub>2</sub>	N <sub>2</sub>	Ar	Cl <sub>2</sub>	WF <sub>6</sub>	CH <sub>3</sub> OH	C <sub>2</sub> H <sub>5</sub> OH			
CM500L	4340 steel	500	-	-	-	75-	-	0.5	1.354	725	60-130
		750	10	-	-	100	-	-	-	-	-
90Ta-10W	4340 steel	840	-	1000	75	-	-	-	0.178-0.445	1425	120-270*
		1040	-	1500	135	-	-	-	-	-	40*
90Ta-10W	Mo	100	-	75	25	-	-	-	0.036-0.0534	1300-1450	60
		400	-	250	100	-	-	-	-	-	-
CM 500	Mo	295	860	-	-	250	120	-	0.926	1165-	30-75
		350	1135	-	-	400	-	-	-	1245	-

\* Includes 30 min TiCl<sub>4</sub> for initial coating of Ti on steel. TiCl<sub>4</sub> turned off after 30 min and Cl<sub>2</sub> flow started through 90Ta-10W alloy chip pot.

Table 2. M30 composition and physico-chemico properties

<u>Composition</u>	<u>Pressure (MPa)</u>
Nitrocellulose (12.6% N)	28.00
Nitroglycerine	22.50
Nitroguanidine	47.7
Ethyl-centralite	1.50
Graphite	1.10
Cryolite	0.30
Ethanol (residual)	0.30
Water (residual)	0.00

<u>Properties*</u>	
T <sub>f</sub> (K)	2990.0
C <sub>p</sub> J/mol-K	43.6
I <sup>p</sup> (J/g)	1072.0
CO (mol/kg)	11.9
H <sub>2</sub> (mol/kg)	5.8
H <sub>2</sub> O (mol/kg)	10.4
N <sub>2</sub> (mol/kg)	11.9
CO <sub>2</sub> (mol/kg)	3.0
Total (mol/kg)	43.1
M <sub>w</sub> (g/g-mol)	22.3
HEX <sub>obs</sub> cal/g	974.0

\* Calculated by Blake Internal Ballistic Code

Table 3. Erosion data for Q4500L coated steel sleeves\*

<u>Pressure</u> (MPa)	<u>Erosion</u> (cm <sup>3</sup> /shot)	<u>Total erosion</u> (cm <sup>3</sup> )	<u>Coating left</u> (%)
<u>Sleeve no. 398 (0.010 cm coating)</u>			
157.78	0.00223	0.00669	89.00
156.06	0.00168	0.00504	80.72
159.37	0.00142	0.00426	73.71
Average	156.26±3.15	0.00178±.00041	
<u>Sleeve no. 400 (0.015 cm coating)</u>			
153.92	0.03003	0.09009	0.78
155.02	0.00082	0.00246	0.00
159.85	0.00049	0.00147	0.00
Average	156.26±3.15	0.0104±.01696	

\* Three successive shots fired in each test.

Table 4. Erosion data for AISI 4340 steel\*

<u>Pressure</u> (MPa)	<u>Average erosion</u> (cm <sup>3</sup> /shot)
102±2	0.00041±.00015
150±1	0.00067±.00014
183±1	0.00106±.00013
260±2	0.00625±.00023

\* Average value based on ten shots.



Table 5. Erosion data for Ta/W coated steel sleeves

<u>No. of shots</u>	<u>Pressure (MPa)</u>	<u>Erosion (cm<sup>3</sup>/shot)</u>	<u>Total erosion (cm<sup>3</sup>)</u>	<u>Coating left (%)</u>
<u>Sleeve no. 107 (0.010 cm coating)</u>				
1	185.17	0.00138	0.00138	99.77
3	183.96	0.02835	0.07505	0.00
Average	184.57±.86	0.01487±0.01907		
<u>Sleeve no. 110 (0.019 cm coating)</u>				
1	183.96	0.01945	0.01945	83.01
3	193.96	0.04269	0.12807	0.00
Average	183.96±.00	0.03107±0.01643		

Table 6. Erosion data for CM500 coated molybdenum sleeves\*

<u>Pressure</u> (MPa)	<u>Erosion</u> (cm <sup>3</sup> /shot)	<u>Coating left</u> (%)	<u>Total cracks</u>
<u>Sleeve no. 1 (0.0355 cm coating)</u>			
144.1	0.000309	99.581	0
183.3	0.001098	98.100	0
220.5	0.001325	96.312	0
<u>Sleeve no. 2 (0.03912 cm coating)</u>			
143.8	0.001567	97.809	0
147.9	0.00149	95.725	0
145.0	0.00053	94.981	0
(avg 0.00149±.000378)			
<u>Sleeve no. 3 (0.03912 cm coating)</u>			
148.8	0.000964	98.740	Split in Half
148.7	0.005093	92.080	
(avg 0.00303±.00292)			
<u>Sleeve no. 4 (0.03912 cm coating)</u>			
145.1	0.001541	98.162	1
144.0	0.002108	95.647	
143.9	0.000459	95.079	
(avg 0.00137±.00084)			
<u>Sleeve no. 5 (0.0521 cm coating)</u>			
146.3	0.002515	97.599	3
144.4	0.004351	93.422	
(avg 0.00343±.00130)			

Table 6. (cont)

<u>Pressure</u> (MPa)	<u>Erosion</u> (cm <sup>3</sup> /shot)	<u>Coating left</u> (%)	<u>Total cracks</u>
<u>Sleeve no. 6 (0.0376 cm coating)</u>			
146.3	0.004206	94.567	3
144.4	0.000686	93.680	
(avg 0.00245±.00249)			
<u>Sleeve no. 7 (0.0287 cm coating)</u>			
144.8	0.00582	99.024	2
179.6	0.000325	98.479	
<u>Sleeve no. 8 (0.1829 cm coating)</u>			
146.3	0.000504	97.907	
143.7	0.002036	92.608	
(avg 0.00142±.00087)			
178.45	0.000428	91.506	3

\* Average value after three shots.

Table 7. Erosion data for Ta/W coated molybdenum sleeves\*

<u>Pressure</u> (MPa)	<u>Erosion</u> (cm <sup>3</sup> /shot)	<u>Coating left</u> (%)	<u>Total cracks</u>
<u>Sleeve no. 9 (0.00381 cm coating)</u>			
140.5	0.00000	100.00	3
144.7	0.00045	98.078	
141.9	0.000379	96.455	
141.2	0.000367	94.874	
(avg 0.00030±.00020)			
184.7	0.000843	91.286	3
185.2	0.000837	87.698	
182.1	0.00109	83.698	
(avg 0.00117±.00051)			
219.1	0.00191	74.838	3
<u>Sample no. 12 (0.01586 cm coating)</u>			
150.2	0.000240	99.750	3
151.0	0.000129	99.616	
151.0	0.000325	99.252	
(avg 0.00014±.000093)			
184.7	0.0009160	93.43	3
186.0	0.002830	92.97	
186.4	0.002950	91.96	
(avg 0.00223±.00051)			
<u>Sample no. 13 (0.04191 cm coating)</u>			
144.0	0.01300	94.732	3
144.0	0.00042	94.56	
149.5	0.000475	94.36	
141.9	0.000669	94.10	
142.6	0.000722	93.8	
(avg 0.00305±.00552)			
183.3	0.000928	93.43	3
186.0	0.001145	92.97	
186.0	0.00335	91.61	
(avg 0.00181±.00134)			

Table 7. (cont)

<u>Pressure</u> (MPa)	<u>Erosion</u> (cm <sup>3</sup> /shot)	<u>Coating left</u> (%)	<u>Total cracks</u>
<u>Sample no. 15 (0.09459 cm coating)</u>			
144.7	0.000915	99.680	
143.3	0.00134	99.220	
143.3	0.000578	99.020	
143.3	0.000892	98.710	
(avg 0.00093±.00031)			
184.0	0.00349	97.50	
183.3	0.002144	96.76	0
(avg 0.00282±.00095)			
<u>Sample no. 16 (0.01905 cm coating)</u>			
148.1	0.000892	99.22	
148.8	0.00350	96.18	
151.6	0.00245	94.08	
(avg 0.00228±.00131)			
188.8	0.08592	19.36	0
<u>Sample no. 17 (0.0127 cm coating)</u>			
149.7	0.00669	91.54	
143.1	0.00546	84.63	
149.5	0.01145	70.14	4
(avg 0.00787±.00316)			

\* Single shot erosion data.

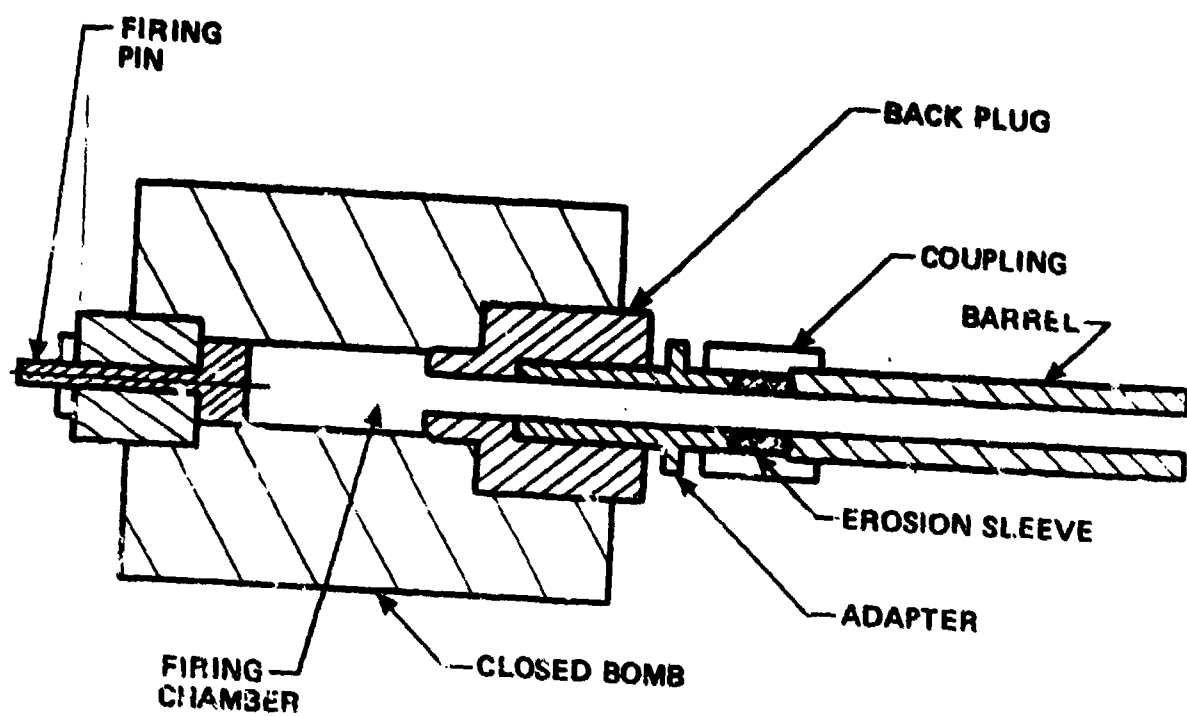


Figure 1. Erosion apparatus



Figure 2. CM500L coating on steel (600X/3000X)



Figure 3. Eroded CM500L coating on steel (550X/2200X)



Figure 4. Fractured and eroded Ta/W coating on steel (150X)



Figure 5. Eroded steel substrate adjacent to Ta/W coating (700X/3500X)





Figure 6. CM500 coating on molybdenum substrate with radial crack



Figure 7. Eroded CM500 coating on molybdenum substrate (60X/350X)



Figure 8. Chipped CM500 coating at bore leading edge (220X/100X)

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